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(54) Title: PLANT CONTROL METHODS

(57) Abstract

Vegetation control is effected by applying to the vegetation a synergistic combination of an amide-sulfuric acid adduct and a quaternary salt. The preferred combination comprises monocarbamide dihydrogen sulfate and diquat.

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PLANT CONTROL METHODS

BACKGROUND OF THE INVENTION

Field of the Invention

The present invention relates to a method for controlling plant growth. The invention particularly relates to a plant control method utilizing the combination of two contact herbicides, which combination is characterized by synergistic systemic activity.

Description of the Art

Control of plants is of special economic importance because of the easier harvesting and accelerated ripening obtainable thereby in agricultural and horticultural crops. Plant control may be accomplished with herbicides. The herbicide may be of either the contact or systemic variety. A contact herbicide requires that a substantial portion of the plant be contacted with the active ingredient of the herbicide being applied. A systemic herbicide translocates through the plant and substantial surface contact is not required.

Chemical agents also are known which are useful for the defoliation of plants, either through hormonal regulation or herbicidal injury, and have been registered with the Environmental Protection Agency for certain selected uses. Their effectiveness frequently varies greatly depending upon the climatic conditions, application rates, and the plant to which they are being applied. Thus, there is need for improved agents for effectively controlling plants to facilitate harvest, for the destruction of plants for weed control, and the like.

SUMMARY OF THE INVENTION

According to one aspect of the invention, there is provided a systemic herbicide composition for controlling vegetation comprising the combination of an amide-sulfuric acid adduct and a quaternary salt, and the combination comprises at least 70 weight percent of the composition.

According to another aspect of the invention, there is provided a composition for controlling vegetation comprising at least 70 weight percent diquat and monocarbamide dihydrogen sulfate.

According to yet another aspect of the invention, there is provided a method of controlling vegetation comprising applying to the vegetation an effective amount of composition comprising a quaternary salt and an amide-sulfuric acid adduct, and the adduct is applied in an amount of about 0.6 to 91 pounds per acre.

The present invention resides in the discovery that a combination of two distinct types of contact herbicides exhibit substantially enhanced systemic plant control activity. The combination also has been found to be more effective than when they are applied individually. This latter result often is termed potentiation or synergism since the combination demonstrates a potentency or activity level exceeding that which would be expected based on knowledge of the potencies of the individual components.

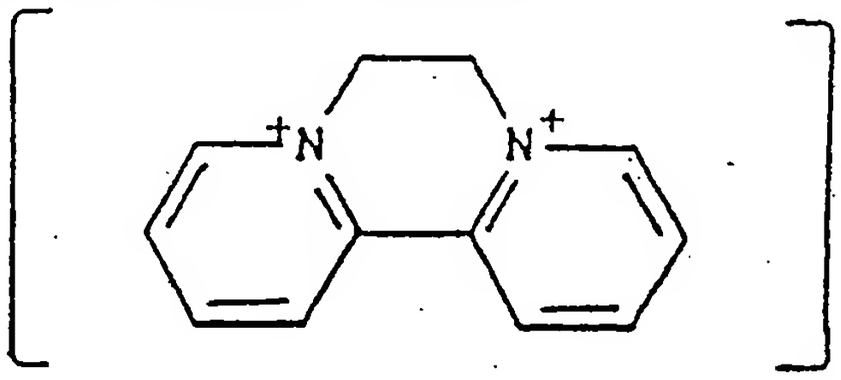
One of the herbicides is a quaternary salt. The other component of the present invention is an amide-sulfuric acid adduct. A surprising aspect of the present invention is that the claimed combination of agents appears to act in a synergistic manner to effect plant control. In one very important discovery, it was found that the quaternary salt and adduct could be used in combination in individual low concentrations (or in individual low application rates) that would render each essentially ineffective for its intended purpose--yet produce, through the combination, a highly efficacious herbicide agent. More specifically, when the quaternary salt is applied at a rate ineffective by itself for significant herbicidal activity in combination with the adduct applied at a rate ineffective by itself for significant herbicidal activity, the result may be a herbicide of high potency.

A particularly preferred composition of the present invention is a combination of diquat and monocarbamide dihydrogen sulfate. The preferred combination has been found to be particularly effective for the dessication of plants.

A particularly surprising aspect of the present invention is that the claimed combination of herbicides appears to act in both a synergistic and highly systemic manner to effect plant control, for example, by dessication. The reason the high systemic activity is particularly surprising is each of the two components are generally considered to be contact agents. More particularly, the quaternary salt and the adduct are generally used as contact herbicides. They generally are used to destroy plants by dessication of the leaves which they contact. When combined in accordance with the present invention, they may produce substantial translocated injury to the plant. Thus, not only is the amount required less than would be expected, the apparent systemic effect is entirely unexpected.

DETAILED DESCRIPTION OF THE INVENTION

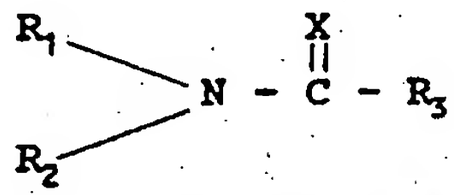
It now has been found that synergism in the control of plants is exhibited by a combination of a quaternary salt and an adduct produced by reaction of sulfuric acid and an amide. The quaternary salts of the present invention are characterized by the general formula:



in which X stands for an anionic radical. The expression "anionic radical" includes such radicals derived from inorganic and organic acids. The anionic radicals may be interchanged one with another without destroying the herbicidal activity of the salt. Suitable anionic radicals derived from inorganic acids are halide radicals, for example, chloride, bromide and iodide radicals. In addition, the anionic radical may be derived from an organic acid such as the P-toluene sulphonate radical. A particularly preferred quaternary salt based on its commercial availability, is

diquat (1,1'-ethylene-2,2'-bipyridylium dibromide) which is commercially formulated as the dibromide monohydrate salt.

The adducts for use in accordance with the present invention are produced by reacting, under controlled conditions, sulfuric acid with an amide having the following formula:



(2)

wherein X is a chalcogen, each of R_1, R_2 and R_3 is independently selected from hydrogen and monovalent organic radicals, and R_1 and R_2, together, can form a divalent organic radical. The molar ratio of amide to sulfuric acid is within the range of about 1/4 to less than 2 so that at least some of the sulfuric acid is present as the monoamide-sulfuric acid adduct. As used herein, "amide" includes all components of formula (2) regardless of the chalcogen employed.

When R_1, R_2 and R_3 are organic radicals, they may be cyclic or acyclic, straight or branched chained and can contain one or more hetero atoms such as sulfur, nitrogen, oxygen, phosphorus and the like. Further, they can contain one or more substituents such as thiol, hydroxy, nitro, amino, nitrile, amide, ester and halogen groups and others. Such organic radicals may contain aryl groups, such as aralkyl and alkaryl groups. The preferred organic radicals are free of olefinic or alkynyl unsaturation and generally have up to about 20, preferably up to about 10 carbon atoms. Particularly preferred amides are urea, thiourea, formamide, biuret, triuret, thioformamide, ethyl formamide, methyl formamide, dimethyl formamide and combinations of these.

The chalcogens are elements of Periodic Group VI-B and include oxygen, sulfur, selenium, tellurium, and polonium. Oxygen and sulfur are presently preferred due to low cost, availability, low toxicity and chemical activity, with oxygen the most preferred. Based upon current test data, the monoadduct is most preferred species.

The combination of the present invention is used advantageously for the control of vegetation. The efficacy

for growth control depends, among other things, on the amount of the combination applied per acre, the relative proportions of adduct to quanternary salt, the treatment time, and the type of plant to which it is applied. The inhibiting effects may occur in a manner which produces, for example, total inhibition of development (destruction) of a waste blend floral including shrubbery. The combination of agents of the invention also can be used wherever it is desired not to fully destroy a floral at once, but to maintain it in a vegetative low growth stage.

Illustrative of vegetation that can be controlled by these methods, with or without the use of surfactants are: black mustard (brassica nigra), curly dock (rumex crispus), common groundsel (senecio vulgaris), pineapple weed (matricaria matricarioides), swamp smartweed (kelp) (polygonum coccineum), prickly lettus (lactuca scariola), lance-leaved groundcherry (physalis lanceifolia), annual sowthistle (sonchus oleraceus), london rocket (sisymbrium irio), common fiddleneck (amsinckia intermedia), hairy nightshade (solanum sarrachoides), shepherd's purse (capsella bursa-pastoris), sunflower (helianthus annus), common knotweed (polygonum aviculare), green amaranth (amaranthus hybridus), mare's tail (conyza canadensis), henbit (lamium amplexicaule), cocklebur (xanthium strumarium), cheeseweed (malva parviflora), lambsquarters (chenopodium album), puncture vine (tribulus terrestris), common purslane (portulaca oleracea), prostrate spurge (euphorbia supina), telegraph plant (heterotheca grandiflora), carpetweed (mollugo verticillata), yellow starthistle (centaurea solstitialis), milk thistle (silybum marianum), mayweed (anthemis cotula), burning nettle (urtica urens), fathen (atriplex patula), chickweed (stellaria media), scarlet pimpernel (anagallis arvensis) redroot pigweed (amaranthus retroflexus), minnerslettuce (montia perfoliata), turkey mullein (eremocarpus setigerus), nettleleaf goosefoot (chenopodium murale), prostrate pigweed (amaranthus blitoides), silverleaf nightshade (solanum elaeagnifolium), hoary cress (cardaria draba), largeseed dodder (cuscuta indecora), California burclover (medicago polymorpha), horse purslane (trianthema portulacastrum), field bindweed

(Iconvolvulus arvensis), Russian knapweed (centaurea repens), flax-leaved fleabane (conyza bonariensis), wild radish (raphanus sativus), tumble pigweed (amaranthus albus), stephanomeria (stephanomeria exigua), wild turnip (brassica campestris), buffalo goard (cucurbita foetidissima), common mullein (verbascum thapsus), dandelion (taraxacum officinale), Spanish thistle (xanthium spinosum), chicory (cichorium intybus), sweet anise (foeniculum vulgare), annual yellow sweetclover (melilotus indica), poison hemlock (conium maculatum), broadleaf filaree (erodium botrys), whitestem filaree (erodium moschatum), redstem filaree (erodium cicutarium), ivyleaf morning-glory (ipomea hederacea), shortpod mustard (brassica geniculata), buckhorn plantain (plantago lacenolata), sticky chickweed (cerastium viscosum), himalaya blackberry (rubus procerus), purslane speedwell (veronica peregrina), Mexican tea (chenopodium ambrosioides), Spanish clover (lotus purshianus), Australian brassbuttons (cotula australis), goldenrod (solidago californica), citron (citrullus lanatus), hedge mustard (sisymbrium orientale), black nightshade (solanum nodiflorum), Chinese thornapple (datura ferox), bristly ox tongue (picris echioides), bull thistle (cirsium vulgare), spiny sowthistle (sonchus asper), Tasmanian goosefoot (chenopodium pumilio), goosefoot (chenopodium botrys), wright groundcherry (physalis acutifolia), tomatillo groundcherry (physalis philadelphica), pretty spurge (euphorbia peplus), bitter apple (cucumis myriocarpus), indian tobacco (nicotiana bigelovii), common morning-glory (ipomoea purpurea), waterplantain (alisma triviale), smartweed (polygonum lapathifolium), mature sowthistle (sonchus asper), yellow nutsedge (cyperus esculentus), purple nutsedge (cyperus rotundus), lupine (lupinus formosus), and grasses of the family Gramineae such as annual rye grass, blue grass, water grass, barnyard grass, bermuda grass, fescue, mat grass, Johnson grass, and the like.

The amide-sulfuric acid adduct can be produced by the reaction of the amide with sulfuric acid by the methods described in U.S. Patent 4,445,925, the disclosure of which is incorporated in its entirety by reference.

While the combination of the present invention can be used alone, it generally is applied to plants in conjunction with other substances, such as a carrier vehicle, wetting agents, emulsifiers, solvents and the like. Suitable carrier vehicles include water, aliphatic or aromatic hydrocarbons, such as benzene, toluene, xylene, cyclohexonone, isophorone, and mineral or vegetable oil fractions. The particularly preferred carrier vehicle is water, based on its availability and cost. Alternatively, of course, a solid carrier vehicle could be utilized. Examples of solid carrier vehicles are minerals, such as siliceous clay, silica gel, talc, kaolin, limestone, and plant products, such as flours.

Typical surface active substances, which may be utilized include calcium-lignin sulfonate, polyoxyethyleneoctylphenol ether and naphthalene-sulfonic acids and their salts, phenolsulfonic acids and their salts, formaldehyde condensates, fatty alcohol sulfates, and substituted benzenesulfonic acids and their salts.

The concentrations of the active agents in the composition of the present invention may vary within wide limits. Typically, a commercial blend of the combination, for sale to a retail outlet or end user, would comprise at least 60, generally in excess of 70 and preferably at least 85 weight percent of the active agents. For field application to plants the composition may be diluted to contain from about 1 to 80 weight percent of the active agents, about 20 to 90 weight percent of a solid or liquid carrier vehicle, and optionally up to 20 weight percent of a surface-active substance.

The ratio of adduct to quaternary salt also may vary widely, although the lower ratio prodive a greater systemic effect. The quaternary salts, for use in the present invention, are typically in the form of powder. The commercial forms generally are in a liquid carrier, for example, 3.73 pounds of the herbicidal salt (comprising two pounds of the active cation ingredient) in a gallon of carrier liquid. One of the preferred adducts is commercially available as ENQUIK from the Union Oil Company of California.

ENQUIK is an aqueous liquid containing approximately 82 weight percent monocarbamide dihydrogen sulfate (MCDS). The ratios are expressed in terms of pounds of the adduct to pounds of active cation in the quaternary salt. Broadly, the ratio of adduct to active cation may be from as low as 1:1 to as high as 1000:1. More typically, the ratio will be within the range of from about 1:1 to 400:1, with a preferred range being about 1:1 to 200:1. The higher ratios are less preferred since they tend to diminish the translocated injury necessary to ensure root kill and complete plant necrosis.

The foregoing ratios are applicable to the preferred compounds and others having similar molecular weights. When compounds having significantly different molecular weights are used, the ratio should be based on their molecular weights. The mole ratio should be about 1:1 to 1160:1, preferably 1:1 to 424:1, and more preferably from 1:1 to 232:1.

The combination of the present invention has been found to be highly effective for control of plants, such as cotton, when the ratio of the adduct to quaternary salt is selected such that, when applied, the amount of adduct applied per acre is within the range of from about 0.6 to 91 pounds, preferably from 1 to 31 pounds, and more preferably within the range of from about 2 to 10 pounds per acre. In a similar manner, the combination is most effective when applied in an amount to provide from about 0.05 to 0.5 pound, preferably from 0.05 to 0.25 pound, and more preferably about 0.1 to 0.2 pound of the active cation constituent of the salt per acre.

To facilitate distribution of a uniform amount of the composition of the invention on plants to be treated, it generally is applied in a carrier vehicle. Water is the preferred carrier vehicle. When the composition is sprayed from the ground, it may be diluted with water to provide a spray volume of from as low as 1 to as high as 200 gallons per acre. Typically, a spray volume of about 10 to 50 gallons per acre is utilized. In the case of aerial spraying, a more concentrated solution is used and is applied at the rate of from about 2 to 15, and more generally 3 to 5 gallons per acre.

A unique aspect of the present invention is that 100 percent coverage of the plants is not required since the combination of the invention exhibits a pronounced systemic effect. Thus, less than 100 percent plant coverage is required. Generally less than 80 percent and frequently less than 60 percent coverage is adequate to effect complete dessication and necrosis of the treated plant. Indeed, as will be seen from the following example, as little as about 10 percent coverage produced substantially complete dessication of the plants treated with a composition of the present invention.

The following example and test data therein are for illustrative purposes only and should not be construed as limiting the scope of the invention.

Example

To demonstrate the efficacy of the present invention, a series of field tests was performed. The quaternary salt utilized was diquat (1,1'-ethylene-2,2'bipyrdylium dibromide), sold commercially in an aqueous carrier and containing approximately two pounds of the active ingredient per gallon of carrier. It is sold for use as a contact herbicide and dessicant for dessication of potato vines, seed crops; control of sugar cane flowering and weed control. It is registered for such purposes with the Environmental Protection Agency. The adduct used was a commercially available monocarbamide dihydrogen sulfate (MCDS) herbicide and dessicant registered with the Environmental Protection Agency for use on a variety of plants. It is sold in a liquid form as ENQUIK, supra, and comprises about 82 weight percent of the active ingredient.

Each herbicide was applied alone and in various combinations to young cotton plants. The plants were approximately eight to twelve inches tall. Each plant had from about 6 to 8 leaves. In each instance, the herbicides were diluted with water to provide the equivalent of a spray volume of approximately 20 gallons per sprayed acre. Each test plot consisted of one plant. A randomized block design

with four replicates was utilized (except plot 5 in which only three replicates were obtained).

The herbicide was applied by hand using a paint brush to apply the herbicide to one fully expanded leaf on each plot (plant). Brushes were changed between each treatment and care was taken to treat only the top surface of the leaf selected for treatment. Each treatment was prepared in a solution which would be equivalent to 20 gallons per acre total spray volume.

Visual ratings of the percent necrosis (dessication) were made at 18, 42, and 70 hours after treatment (HAT). Immediately following the final rating, the plants were harvested and fresh weights were taken. Plants were then allowed to air dry until their weight had stabilized for three consecutive days. Dry weights were then taken. The percent weight change was calculated by taking the fresh weight divided by the dry weight and then multiplying by 100. The results of these tests are set forth in the table below.

TABLE

Plot	18 HAT % Neor.	42 HAT % Neor.	70 HAT % Neor.	Fresh	Dry	% Change	Equivalent Herbicide Application Rate per acre ENQUIK	diquat
1	0	10	10	20.79	5.13	405	3 Gal.	0
	0	10	10	10.54	2.57	510		
	0	10	10	8.55	2.33	367		
	0	10	10	9.10	2.41	378		
Average	0	10	10	12.25	3.11	390		
2	0	10	10	17.27	3.91	442	5 Gal.	0
	0	10	10	13.26	3.01	441		
	0	10	10	22.70	4.73	480		
	0	10	10	14.65	3.61	406		
Average	0	10	10	16.97	3.82	442		
3	0	15	15	17.49	3.84	455	0 Gal.	8 oz.
	0	15	15	8.14	1.98	411		
	0	15	15	10.83	2.51	431		
	0	15	15	11.41	3.00	380		
Average	0	15	15	11.96	2.83	419		
4	20	30	35	12.41	3.32	374	0	16 oz.
	0	15	15	8.99	2.70	333		
	0	20	20	11.23	2.85	394		
	0	15	15	8.78	2.60	338		
Average	5	20	21	10.35	2.87	360		
5	0	30	40	15.78	4.78	330	0	32 oz.
	0	40	60	6.50	2.60	250		
	5	25	30	10.13	3.27	310		
Average	2	32	43	10.80	3.55	297		

Table (continued)

Plot	18 HAT % Neor.	42 HAT % Neor.	70 HAT % Neor.	Fresh	Dry	% Change	Equivalent Herbicide Application Rate per acre ENQUIK	diquat*
6	5	40	50	9.51	2.94	311	3 Gal.	8 oz.
	5	60	85	4.98	2.42	206		
	5	50	100	5.50	3.07	179		
	5	50	90	4.80	2.39	201		
Average	5	50	81	6.11	2.71	224		
7	20	90	100	4.38	2.67	164	3 Gal.	16 oz.
	15	90	100	3.59	2.37	151		
	15	60	85	5.82	2.61	223		
	30	95	100	3.53	2.25	157		
Average	20	84	96	4.33	2.48	174		
8	20	98	100	2.79	2.05	136	3 Gal.	32 oz.
	10	90	100	3.76	2.62	144		
	40	98	100	3.28	2.34	140		
	35	100	100	4.58	2.91	257		
Average	26	97	100	3.60	2.48	144		
9	30	75	100	7.50	3.53	212	5 Gal.	8 oz.
	15	70	98	3.39	1.88	180		
	5	80	100	2.71	1.54	176		
Average	17	75	99	4.53	2.32	189		
10	10	80	95	4.48	2.33	192	5 Gal.	16 oz.
	40	70	80	6.75	2.69	251		
	35	95	100	3.07	2.00	154		
	20	85	100	2.72	1.65	165		
Average	26	83	94	4.26	2.17	191		

Table (continued)

Plot	18 HAT % Neor.	42 HAT % Neor.	70 HAT % Neor.	Fresh	Dry	% Change	Equivalent Herbicide Application Rate per acre ENQUIK	diquat*
11	50	98	100	3.32	2.13	156	5 Gal.	32 oz.
	10	75	100	5.20	3.22	161		
	15	98	100	5.47	3.35	163		
	15	50	45	15.88	5.63	282		
Average	30	80	86	7.47	3.58	191		

% Neor. = Percent necrosis

Fresh = Fresh weight in grams

Dry = Dry weight in grams

% Change Percent weight change [(Fresh/Dry) X 100]

* Liquid volume of a commercial formulation containing 2 lbs. of active ingredient per gallon of carrier fluid

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From the table it is seen that, 18 hours after treatment (HAT), treatments of MCDS (ENQUIK) diquat alone, and MCDS plus diquat (3 gallons plus 8 ounces) caused injury only to the treated leaf (5 percent or less necrosis). All other mixtures of MCDS and diquat caused noticeable translocated injury (from 17 to 30 percent necrosis).

At 42 hours after treatment, the effects of addition of MCDS to diquat were clearly visible with increases in necrosis of from 150 percent to 477 percent compared to equivalent rates of diquat alone. With all combinations of MCDS and diqaut, injury was apparent on leaves both above and below the treated leaf indicating translocated injury (mobility of the herbicide through the plant system).

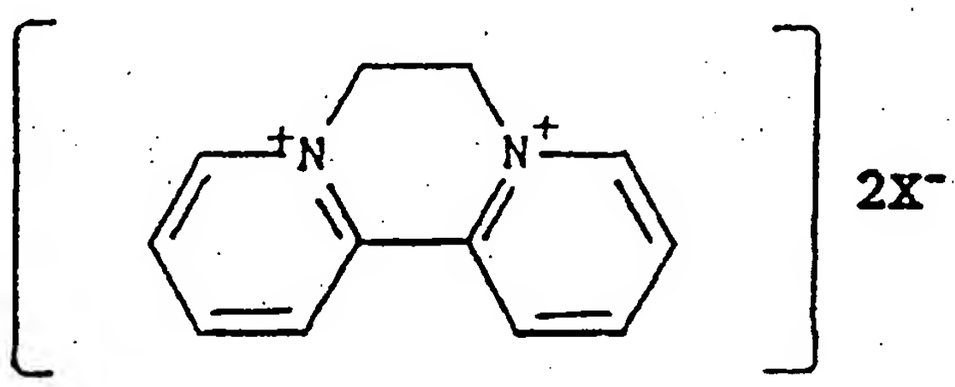
Similarly, at 70 hours after treatment, necrosis was increased by from 95 percent to 661 percent with mixtures of MCDS and diquat compared to diquat alone or MCDS alone. Further, analysis of the percent weight change data provided results similar to the necrosis data. This further verifies the conclusion that the combination of MCDS and diquat is not only synergistic in the response produced, but that the combination of two basically contact herbicides produces a mixture which behaves in a systemic manner (translocates).

While particular embodiments of the invention have been described, it will be understood that the invention is not limited thereto. Many modifications can be made. For example, while the invention has been described with respect to the two particularly preferred compounds, and their use on cotton, it will be readily apparent to those skilled in the art that other combinations within the scope of the invention can be utilized on cotton and on other plants as well. It is intended that such modifications as will fall within the spirit and scope of the appended claims are included.

C L A I M S

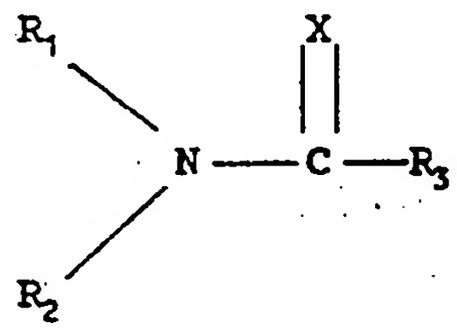
1. A systemic herbicide composition for controlling vegetation comprising the combination of an amide-sulfuric acid adduct and a quaternary salt, and said combination comprises at least 70 weight percent of the composition.

2. The composition of Claim 1 wherein the quaternary salt has the formula:



wherein X is an anion.

3. The composition of Claim 2 wherein the amide has the formula:

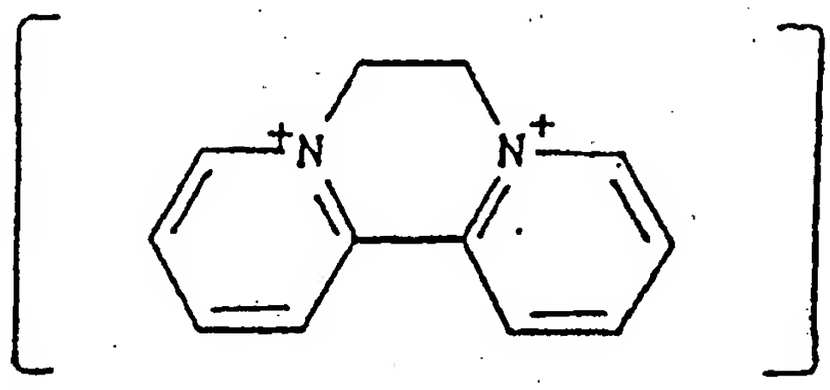


wherein X is a chalcogen each of $R_{1, R2}$ and R_3 is independently selected from hydrogen and monovalent organic radicals, and R_1 and R_2 together can form a divalent organic radical.

4. The composition of Claim 1 wherein the adduct and salt are present in amounts to provide a weight ratio of from 1:1 to 400:1 based on the active ingredients.

5. A composition for controlling vegetation comprising at least 70 weight percent diquat and monocarbamide dihydrogen sulfate.

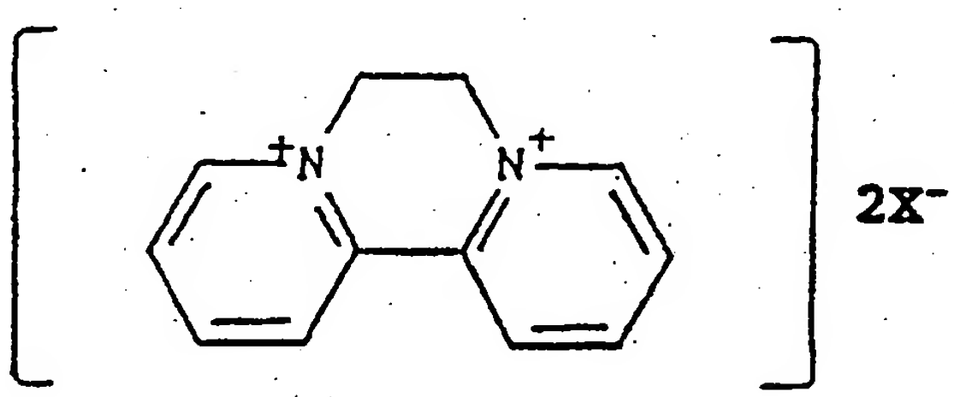
6. The composition of claim 1 wherein the quanternary salt is a compound of the formula



wherein X is a halide ion and said adduct comprises a monoamide adduct of sulfuric acid.

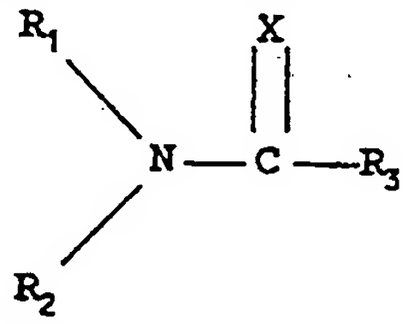
7. The composition of claim 4 wherein the salt and adduct are present in an amount to provide a weight ratio of adduct to salt within the range of from about 1:1 to 400:1 based on the active ingredients.

8. The composition of claim 1 wherein the adduct is monocarbamide dihydrogen-sulfate and the salt is a compound of the formula



wherein X is an anion.

9. The composition of claim 1 wherein the amide is a composition having the formula



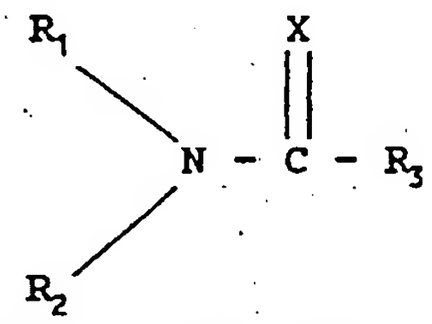
wherein X is a chalcogen, each of R_1, R_2 and R_3, is independently selected from hydrogen and monovalent organic

radicals, and R_1 and R_2 together can form a divalent organic radical.

10. The composition of Claim 1 wherein the salt comprises diquat.

11. The composition of Claim 1 wherein the adduct is monocarbamide dihydrogen-sulfate.

12. The composition of Claim 1 wherein the quaternary salt is diquat and the amide is a composition having the formula



wherein X is a chalcogen, each of R_1, R_2 and R_3, is independently selected from hydrogen and monovalent organic radicals, and R_1 and R_2 together can form a divalent organic radical.

13. The composition of Claim 1 wherein the adduct is the reaction product of sulfuric acid and an amide selected from the group consisting of urea, thiourea, formamide, biuret, triuret, thioformamide, ethyl formamide, methyl formamide, dimethyl formamide and combinations thereof.

14. The composition of Claim 6 wherein the adduct is the reaction product of sulfuric acid and an amide selected from the group consisting of urea, thiourea, formamide, biuret, triuret, thioformamide, ethyl formamide, methyl formamide, dimethyl formamide and combinations thereof.

15. A composition for controlling vegetation comprising the reaction product of sulfuric acid and an amide selected from the group consisting of urea, thiourea, formamide, biuret, triuret, thioformamide, ethyl formamide, methyl formamide, dimethyl formamide, and combinations

thereof, and diquat said compositiojn having a weight ratio of reaction product to active cation of diquat of 1:1 to 400:1.

16. The composition of claim 15 wherein the diquat and adduct are present in an amount to provide a weight ratio of adduct to diquat within the range of from about 1:1 to 200:1 based on the active ingredients.

17. The composition of Claim 16 wherein the diquat and adduct comprise at least 85 weight percent of the composition, and the composition further includes a carrier vehicle.

18. The composition of Claim 17 wherein the carrier vehicle is water.

19. A method of controlling vegetation comprising applying to the vegetation an effective amount of composition comprising a quaternary salt and an amide-sulfuric acid adduct, and said adduct is applied in an amount of about 0.6 to 91 pounds per acre.

20. The method of Claim 19 wherein the composition is applied in an amount to provide from about 0.05 to .26 pound per acre of the active cation of the salt.

21. The method of Claim 20 wherein the composition is diluted with sufficient water to provide a spray volume of from 10 to 50 gallons per acre.

22. The method of Claim 20 wherein the composition is applied in an effective amount to dessicate the vegetation.

23. The method of Claim 20 wherein the salt is diquat and the adduct is monocarbamide dihydrogen sulfate.

24. The method of Claim 20 wherein the adduct and salt are present in an amount to provide a weight ratio of

adduct to salt within the range of from about 1:1 to 400:1 based on the active ingredient of the salt.

25. The method of claim 19 wherein the adduct and salt are each contact herbicides, and the adduct comprises a monoamide adduct of sulfuric acid.

26. The method of claim 25 wherein the adduct and salt, are present in an amount to provide a weight ratio of adduct to salt in the range of from about 1:1 to 200:1 based on the active ingredient of the salt.

27. A method of producing a herbicide comprising admixing a quanternary salt with an adduct reaction product of an amide and sulfuric acid in amounts to provide a weight ratio of adduct to active cation of the salt of from 1:1 to 400:1.

28. The method of Claim 27 wherein the quaternary salt and adduct are admixed in the presence of a liquid carrier.

29. The method of Claim 28 wherein the liquid carrier is water.

30. The method of Claim 29 wherein the quaternary salt is diquat and the adduct is monocarbamide dihydrogen sulfate.

31. The method of Claim 30 wherein the amounts of monocarbamide dihydrogen sulfate and diquat provide a weight ratio of monocarbamide dihydrogen sulfate to diquat within the range from about 1:1 to 200:1 based on the active ingredients.

32. A composition produced by the method of Claim 28.

33. A composition produced by the method of Claim 31.

34. A method of controlling vegetation comprising applying to the vegetation a herbicidally effective amount of a combination of a quaternary salt and an amide-sulfuric acid adduct, said combination having a weight ratio of adduct to active ingredient of the quaternary salt of 1:1 to 400:1.

INTERNATIONAL SEARCH REPORT

International Application No PCT/US 93/00128

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all)[6]	
According to International Patent Classification (IPC) or to both National Classification and IPC Int.Cl. 5 A01N59/02; //(A01N59/02,43:90)	
II. FIELDS SEARCHED	
Minimum Documentation Searched[7]	
Classification System	**Classification Symbols**
Int.Cl. 5	A01N
Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched[8]	

III. DOCUMENTS CONSIDERED TO BE RELEVANT[9]

Category °	Citation of Document,[11] with indication, where appropriate, of the relevant passages[12]	Relevant to Claim No.[13]
X	EP,A,0 243 522 (UNION OIL COMPANY OF CALIFORNIA) 4 November 1987 see page 2, line 4 - line 6 see page 3, line 16 - page 14, line 15 see page 15; examples 8,9	1,4,7,9, 11,13, 14, 19-22, 24-29, 32,34
Y		2,3,5,6, 8,10,12, 15-18, 23,30, 31,33

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° Special categories of cited documents :[10]

"A" document defining the general state of the art which is not considered to be of particular relevance

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

IV. CERTIFICATION

Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report
31 MARCH 1993	23. 04. 93
International Searching Authority	**Signature of Authorized Officer**
EUROPEAN PATENT OFFICE	LAMERS W.

III. DOCUMENTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)

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A	CHEMICAL ABSTRACTS, vol. 101, no. 5 Columbus, Ohio, US; abstract no. 34517t, H.BOEHM ET AL. 'Effect of Permeation-Promoting Substances on the Action of Plant Protecting Agents' see abstract & WISS. BEITR. - MARTIN-LUTHER-UNIV. HALLE-WITTENBERG vol. 4, 1984, pages 36 - 46	1-34
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III. DOCUMENTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)

Category °	Citation of Document, with indication, where appropriate, of the relevant passages	Relevant to Claim No.
A	EP,A,0 271 173 (UNION OIL COMPANY OF CALIFORNIA) 15 June 1988 see the whole document -----	1-34

ANNEX TO THE INTERNATIONAL SEARCH REPORT ON INTERNATIONAL PATENT APPLICATION NO.

US 9300128
SA 68920

This annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report.
The members are as contained in the European Patent Office EDP file on
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US 9300128
SA 68920

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The members are as contained in the European Patent Office EDP file on
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